

## Feasibility of Graphene-Enhanced Strength in Sanitary Ceramics

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In the competitive production of sanitary ceramics (SCs), fractures during the drying and handling stages significantly contribute to production waste. This study explores the incorporation of graphene, known for its exceptional mechanical properties, to enhance the strength and reduce waste in SCs. Utilising a modified slip casting process, graphene was introduced into Vitreous China (VC) and Fine Fireclay (FFC) ceramics through two methods representing two points of possible graphene inclusion along the production process: An earlier inclusion stage, which was examined by adding graphene solution into ceramic powder to create slip (powder-solution) and a later stage represented by graphene solution added to a readymade slip (solution-solution). The results demonstrated that the solution method significantly improved the mechanical strength of VC, achieving a 12% increase at 1g graphene inclusion, while the powder method caused a decline in strength due to slip flocculation. FFC showed consistent strength improvements with both methods, with an average increase of 7% at 1g graphene inclusion. Although the potential for graphene-enhanced ceramics is promising, further research is needed to quantify material savings, evaluate cost-effectiveness, and optimise the integration process for industrial-scale production.

**Keywords:** Graphene, Sanitary ceramics, Slip Casting, Vitreous China, Fine Fireclay.

### 1. Introduction

Ceramics manufacturing is one of humanity's oldest and most enduring industries, tracing its origins back to prehistoric times. Over millennia, the processes involved in ceramic production have been refined through the adoption of advanced technologies and innovative practices aimed at enhancing quality and minimising waste. This evolution is particularly evident in the production of sanitary ceramics (SC), a sector characterised by large-scale operations and high production volumes. Due to the complex shapes and fine details required in products, the manufacturing process often encounters challenges such as deformation and microcracks during the drying stage and handling. These defects necessitate substantial rework, which incurs significant costs for manufacturers.

One of the most promising developments in material science in recent years has been the advent of graphene. Since its isolation as a monolayer in 2004, graphene has been the focus of extensive research due to its extraordinary mechanical, electrical, and thermal properties [1]. It has been successfully incorporated into various materials, creating composites that exhibit enhanced mechanical properties [2,3]. However, to the best of the authors' knowledge, graphene has not yet been explored as an additive in sanitary ceramics. This oversight may be due in part to the historically high cost of graphene. However, as the bulk price of graphene has steadily decreased over recent years, it has become more accessible, presenting new opportunities for its application in SC manufacturing [4,5]. The inclusion of graphene in ceramic materials is anticipated to offer several key advantages. Firstly, the crack propagation resistance observed in graphene-reinforced composites, as demonstrated in applications such as concrete and structural ceramics, suggests that graphene could similarly mitigate issues like microcracks in sanitary ceramics [6,7]. This is accomplished primarily by crack-bridging as the pullout of graphene increases energy dissipation [8]. This would reduce the need for rework and enhance the overall efficiency of the manufacturing process. Secondly, graphene's ability to significantly enhance the mechanical performance of the materials into which it is incorporated could enable the production of thinner, yet equally robust, ceramic products due to the higher strength-to-weight ratio [9,10]. This not only allows for more complex and aesthetically pleasing shapes but also reduces the amount of ceramic slip required, thereby lowering material costs. Moreover, the enhanced resilience and handling properties imparted by graphene as well as the antibacterial properties [11] could add substantial value to the final sanitary ceramic products, making them more durable and user-friendly.

Despite the extensive research on graphene's notable properties and its successful application in various composite materials, its integration into sanitary ceramics remains unexplored. This study addresses this research gap by

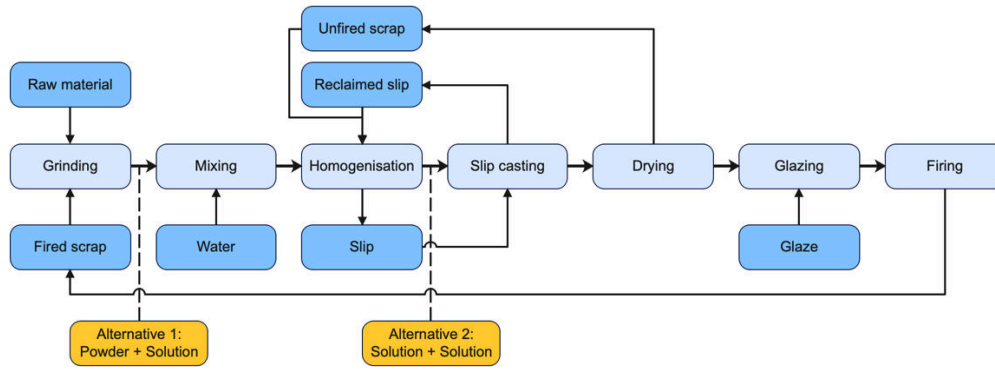
investigating the feasibility and impact of incorporating graphene into sanitary ceramics, specifically focusing on how different methods of graphene introduction affect the mechanical properties of Vitreous China (VC) and Fine Fireclay (FFC), two of the most commonly used clays in the industry. This work contributes to the field by offering a novel approach to enhancing the strength and durability of sanitary ceramics, potentially reducing waste and improving manufacturing efficiency thereby offering tangible benefits to both manufacturers and end-users.

## 2. Manufacturing process of Sanitary Ceramics

The manufacturing process of SCs involves several key stages, with slip casting being a widely utilised technique for producing various types of ceramics. Slip casting involves creating a suspension of ceramic particles, known as slip, within a liquid matrix typically composed of water. This process is integral to forming complex shapes with high precision [12]. The production process, demonstrated in Figure 1, starts with the grinding of raw materials, such as clay, feldspar, and silica. These materials are ground into fine powders to ensure homogeneity in the final product. Following this, the mixing stage involves combining these powders with water to create a consistent slurry. This mixture must be thoroughly blended to ensure the even distribution of all components. Next is the homogenisation of the slip, where the slurry is further refined to achieve a uniform consistency. This stage is crucial as it determines the quality and properties of the slip, influencing the final product's strength and durability. In the slip casting stage, the homogenised slip, which is characterised by its fine grain size, is poured into a porous mould. Through capillary action, the liquid matrix is partially absorbed by the mould, leaving behind a layer of ceramic particles that form the shape of the final product. Once the desired wall thickness is achieved, the excess slip is drained, and the ceramic cast is left to dry before being removed from the mould. The formed products are subsequently dried to remove any remaining moisture. Once dried, the ceramics undergo glazing, where a vitreous coating is applied to the surface to enhance both aesthetics and functionality, such as water resistance. The final step in the process is firing, where the ceramics are heated in kilns at high temperatures, causing the materials to sinter and develop their final mechanical properties [13,14]. Throughout this process, waste management is critical. Unfired scrap and reclaimed slip from the casting process are typically reintroduced into the homogenisation stage to maximise material efficiency and reduce waste. Fired scrap, on the other hand, is recycled by being reintroduced into the grinding stage, ensuring that even the fired waste contributes to the production cycle.

The integration of graphene into the manufacturing process of sanitary ceramics presents both opportunities and challenges. While graphene's exceptional mechanical properties hold the potential to significantly enhance the strength and durability of ceramics, the practical challenges of incorporating it into the existing production processes cannot be overlooked. The primary challenges include ensuring uniform dispersion of graphene within the ceramic matrix, preventing agglomeration, and managing the interaction between graphene and other additives. The high viscosity of ceramic slips, especially when produced on an industrial scale, complicates the even distribution of graphene, which could lead to areas of weakness within the final product. Moreover, graphene's safety considerations like becoming airborne, posing health risks and handling difficulties [15]. Overcoming these challenges requires careful consideration of the stage at which graphene is introduced, as well as the medium used for its dispersion. By addressing these issues, this study aims to provide practical insights into the feasibility of scaling up graphene-enhanced ceramics for industrial production.

Given these challenges, the introduction of graphene into the manufacturing process can be approached in two primary ways (figure 1). The first option involves introducing the graphene solution during the initial mixing stage, where it is gradually combined with the raw powders and water. This method allows the graphene to integrate fully with the ceramic particles from the outset, potentially leading to a more homogenous final product as the graphene is introduced into a less viscous slurry, enabling a gradual increase in viscosity which aids dispersion. Alternatively, graphene solution can be added during the homogenisation stage. By introducing graphene at this later stage, the process may offer better control over the dispersion of graphene within the slip while minimising the need for significant alterations to the existing manufacturing line. The subsequent section will detail the experimental procedures used to evaluate the effectiveness of these two methods for incorporating graphene into SCs focusing on VC and FFC. Known for their distinct properties, VC has high density and low porosity, making it ideal for sanitary applications, and FFC's workability makes it ideal for shaping complex ceramic forms [16]. Both materials will be examined to determine how graphene can enhance their properties and improve the overall production process.



**Figure 1 Manufacturing process of SCs as well as the potential graphene inclusion points**

### 3. Methodology

This section details the materials and procedures of the experimental design used to evaluate the impact of incorporating graphene into SCs. The goal is to assess how graphene influences the mechanical properties of two commonly used ceramic materials: VC and FFC. The systematic methodology was designed to ensure accurate and reliable analysis of the results.

#### 3.1 Materials Used

The graphene used in this study was a multilayer ( $9 \pm 6$ ), non-functionalised graphene grade known as G3. This particular form of graphene was selected primarily due to its cost-effectiveness compared to single-layer graphene, making it more practical for potential industrial-scale applications. Additionally, the choice of non-functionalised graphene helps avoid potential chemical interactions between functional groups and the ceramic matrix, which could complicate the material's integration. The multilayer structure of G3, while slightly less effective in some applications compared to single-layer graphene, still provides significant mechanical reinforcement at a more economical price point, aligning with the project's cost-efficiency goals [17]. The material was supplied by Levidian Nanosystems.

In determining the appropriate graphene loading, three inclusion levels were selected: 0.03, 0.06, and 1 gram per 0.5 litres of slip. These concentrations correspond to approximately 0.06 w/v% to 0.2 w/v%. The chosen range was based on studies in related fields, such as concrete and structural ceramics, where optimal graphene loading typically ranges from 0.05 to 1 wt% [18–21]. Despite the low concentrations, sufficient reinforcement was obtained without significantly altering the material's processing characteristics. This selection aims to balance the potential mechanical benefits of graphene reinforcement with the practical considerations of cost and ease of incorporation into the production process.

One of the significant challenges in this experiment was ensuring the proper distribution of graphene within the ceramic slip, given graphene's hydrophobic nature and the safe handling of the material. Initial attempts to disperse graphene in water were unsuccessful, as the graphene did not mix well due to its poor affinity for water. Ethanol was subsequently chosen as the dispersion medium, proving to be far more effective. Ethanol's ability to dissolve hydrophobic materials and its common use in the literature as a solvent for graphene made it the ideal choice [22–24]. Additionally, ethanol is chemically stable in the context of ceramic slips, minimising the risk of unintended interactions with the slip components [25,26]. The chosen ratio was 1 v/v% of ethanol to slip (5 ml of ethanol per 500 ml of slip), ensuring that the graphene was evenly distributed throughout the ceramic matrix without compromising the slip's integrity.

#### 3.2 Slip Casting Process

The slip-casting process used in this study was adapted from the standard methodology employed by a large-scale ceramic manufacturer, with modifications to accommodate graphene integration (figure 2). Two methods of graphene incorporation were tested:

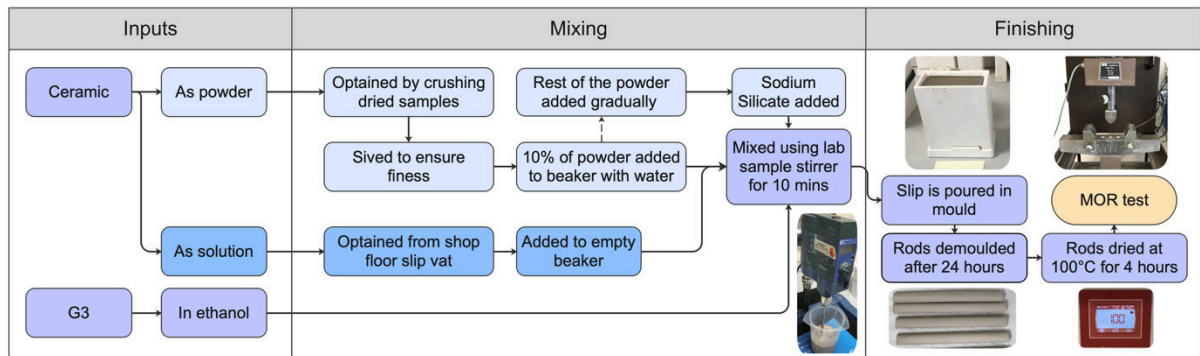
- **Powder-Solution Method:** In this approach, the procedure began with the grinding and sieving of fired ceramic scrap, which was then used to prepare the slip. The graphene-ethanol solution was mixed with the pulverised ceramic powder and water. This method simulates the early-stage introduction of graphene

during the production process, allowing for a gradual increase in viscosity, which is hypothesised to improve the homogeneity of the graphene distribution.

- **Solution-Solution Method:** Here, the graphene-ethanol solution was directly added to the pre-prepared slip from the shop floor. This method represents a later-stage addition, where the graphene solution is mixed into the already homogenised slip. This approach could be more practical for large-scale production, as it simplifies the dosing process.

In both approaches, the mixtures were stirred using a lab mixer, which replicates the mixing conditions found in industrial settings. The mixed slips were then cast into moulds, allowed to rest for 24 hours, and subsequently demoulded. The demoulded parts were dried in an oven at 100°C for four hours to ensure complete moisture removal. Eight specimens were produced for each graphene inclusion level, along with eight baseline specimens for both clay types without graphene to establish a control group.

It is important to note that the slip obtained from the shop floor contained surfactants, while the slip made from pulverised powder was treated with sodium silicate. Sodium silicate, acting as a deflocculant, helps reduce surface tension, thereby improving the integration of the graphene solution into the slip. This property is expected to enhance the uniformity of graphene dispersion and ultimately the mechanical properties of the final product.



**Figure 2 Slip casting process**

### 3.3 Strength Testing

The mechanical properties of the graphene-enhanced ceramics were assessed using the Modulus of Rupture (MOR), which is a standard test for evaluating flexural strength in brittle materials like ceramics. The three-point bending test setup, shown in Figure 3, involved applying force at the centre of each specimen, which was supported at two equidistant points. SCs are brittle materials, and it is generally accepted that the strengths of brittle substances are statistical in nature. Differences in strength from one sample to another occur from variations in defects in the microstructure. Therefore, it is essential to use sample sets with several specimens [27]. This test provides critical insights into the material's bond strength and the uniformity of its microstructure. For context, the standard flexural strength values for VC and FFC are 500±20 PSI and 300±30 PSI, respectively. All specimens, including both the graphene-reinforced and baseline samples, were tested to ensure the reliability and robustness of the results.

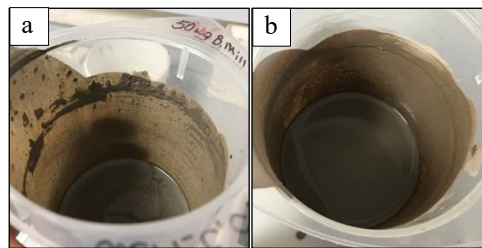


**Figure 3 Hounsfield MOR machine setup**

The methodology outlined has been designed to test the impact of graphene on the mechanical properties of sanitary ceramics. By carefully selecting materials, optimising the dispersion medium, and adapting the slip-casting process, this study aimed to provide valuable insights into the feasibility and potential advantages of incorporating graphene into the production of SCs. The following sections will present and analyse the results, providing an evaluation of the mechanical performance of graphene-enhanced ceramics.

#### 4. Results and discussion

Preliminary inspection of the produced slips while in production displayed no visual signs of agglomerations. However, pouring the slip made using crushed powder into the moulds has revealed flocculated VC material left over on the walls of the beaker as observed in Figure 4.a. However, the graphene slip mixture in which the slip was obtained from the shop floor, as well as the FFC slip showed no signs of flocculation, an example is shown in Figure 4.b. The images were captured with an inclusion concentration of 0.3 grams of G3, nevertheless, similar observations have been made for other concentrations. Slip flocculation usually occurs when the charged colloidal particles cease suspension to sediment under the form of floc. As stated previously, Sodium silicate is added to neutralise the charges of particles in the slip to prevent this. However, G3 inclusion has seemingly caused an unfavourable reaction that negated the effect of sodium silicate on VC slip made from recycling fired scrap and induced flocculation.



**Figure 4 Beakers after the slip is poured into the mould made using the Powder-Solution method for a) VC-G3 and b) FFC-G3**

##### 4.1 Strength results

The average strength values as a function of G3 content were determined for VC and FFC and plotted in Figure 5, with accompanying min-max error bars and comparisons to standard strength values provided by the manufacturer.

##### 4.2 Graphene's Impact on VC (figure 5.a.)

For VC, the baseline strength in the control group was consistent across both methods and aligned with manufacturer standards. However, the method of graphene introduction significantly influenced the results:

- Powder-Solution Method: VC specimens produced from pulverised pieces showed a declining strength as graphene content increased. This reduction in strength can likely be attributed to the flocculation of the slip. The graphene, when introduced early in the process with pulverised ceramic, may have disrupted the dispersion stability, causing particle clumping. This interaction likely interfered with the sodium silicate's role as a deflocculant, which is critical for maintaining a homogenous slip. The resulting heterogeneity in the slip would contribute to weaker points within the ceramic structure, thus reducing overall strength.
- Solution - Solution Method: Conversely, adding graphene directly to the already-produced slip resulted in a notable increase in strength, with a peak improvement of approximately 12% at a 1g inclusion level. This suggests that introducing graphene at a later stage, when the slip's viscosity is more stable, allows for better dispersion of graphene particles. This uniform distribution likely minimises defects and enhances the structural integrity of the ceramic, thereby increasing its strength.

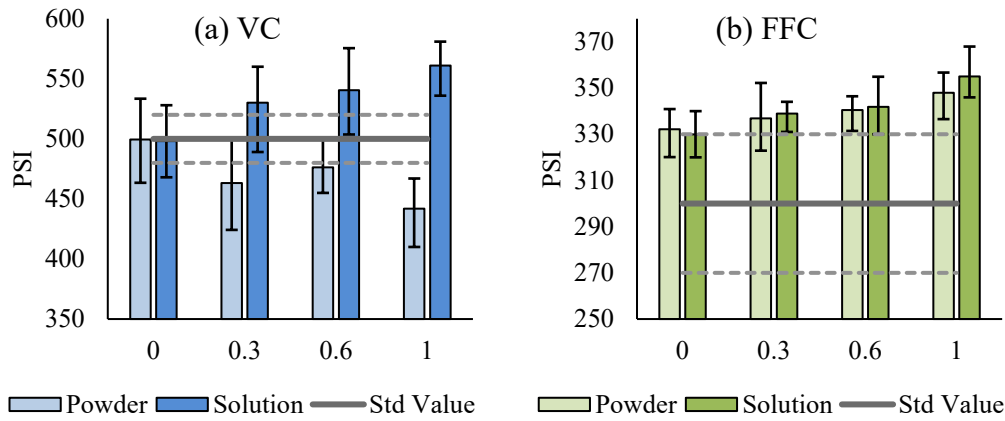
##### 4.3 Graphene's Impact on FFC (figure 5.b.)

In FFC, both methods showed an increase in strength:

- Powder Method: Similar to VC, the FFC specimens exhibited an increase in strength with the introduction of graphene, reaching an approximate improvement of 6%. This increase suggests that

FFC's composition and microstructure might be more compatible with graphene, allowing for better integration even when introduced early in the slip formation process.

- **Solution Method:** The solution method led to a slightly higher increase in strength, around 7.5%. This further supports the hypothesis that better dispersion and control over the introduction of graphene lead to improved mechanical properties. The success of the solution method in both VC and FFC highlights the importance of maintaining uniform graphene dispersion, which is easier to achieve when added to a fully homogenised slip.



**Figure 5 MOR results along with standard strength values for a) VC and b) FFC**

#### 4.4 Summary and Implications

The superior performance of the solution method in this study can be attributed to several key factors. First, the late-stage introduction of graphene in a well-homogenised slip allowed for better control over the dispersion process. The declining strength in VC when using the powder method suggests that graphene can have negative effects if not properly dispersed, likely due to its interaction with slip constituents and the resulting flocculation which allowed the premature increase in viscosity and the subsequent difficulties in achieving a uniform mixture. In contrast, FFC appears more resilient to these effects, possibly due to its different material composition.

These findings underscore the importance of carefully considering the stage at which graphene is introduced during the manufacturing process. The results indicate that the method of graphene introduction significantly influences the mechanical properties of SC ceramics, with the solution method generally yielding better outcomes. Additionally, the use of ethanol as a dispersion medium helped maintain the graphene's stability within the slip, minimising agglomeration and ensuring a more even distribution of particles throughout the ceramic matrix.

The implications of these findings are significant for industrial applications. The solution method not only enhances the mechanical properties of the ceramics but also offers a more straightforward and scalable approach to graphene integration. This is particularly important for large-scale production, where maintaining consistency and quality across batches is critical.

#### 5. Conclusion

In this study, the impact of graphene incorporation on the mechanical properties of sanitary ceramics was systematically evaluated using two types of clay, namely Vitreous China and Fine Fireclay. The results demonstrated that the method of graphene introduction plays a crucial role in determining the final strength of the ceramics. For VC, the solution method led to a significant strength increase, with a peak improvement of 12% at a 1g inclusion level, while the powder method resulted in a decline in strength due to slip flocculation. In contrast, FFC showed consistent strength improvements with both methods, reaching a 7.5% increase with the solution method. These findings suggest that incorporating graphene at a later stage of production is more effective for material integration, particularly in maintaining uniform dispersion and avoiding adverse interactions with slip components. The broader implications of this work indicate that graphene-enhanced sanitary ceramics have the potential to reduce waste, improve product durability, and offer new opportunities for more efficient and cost-effective manufacturing processes.

It is worth noting that while this study focused on the improvement of mechanical strength, graphene has also been shown to enhance fracture toughness in other ceramic systems [28]. Future investigations should explore the potential of graphene to not only increase strength but also improve the fracture toughness of sanitary ceramics, which would further enhance the material's resistance to cracking and mechanical failure during use. Additionally, further research should aim to optimise the integration method by exploring alternative dispersion mediums or additional additives to improve graphene's compatibility with ceramic slips. Moreover, the development of more precise dosing techniques with varying grades of graphene could help refine the process, ensuring that the benefits of the nanoparticles are fully realised without compromising the manufacturability of the ceramics. Moreover, additional studies are needed to quantify potential material savings from the strength increase and evaluate the cost-effectiveness relative to the cost of graphene.

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